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14. ABSTRACT Work on this program was aimed at developing and understanding nano-optical structures with emphasis on developing quantum optical based devices. Specific work focused on semiconductor quantum dots. During this research period, a number of important discoveries were made as well as critical demonstrations of importance to future technology. The discoveries include the prediction and observation of spontaneous emission induced coherence and the unexpected control of nuclear field fluctuations through coherent electron spin trapping that reduced the nuclear fluctuations and increases the electron spin coherence time. Demonstrations include fast spin state initialization, coherent spin trapping, quantum dot tomography, and the Mollow absorption spectrum for neutral and negatively charged exciton. Future work will capitalize on this progress to demonstrate <i>deterministic entanglement</i> between the electron spin and a photon for quantum information transfer and entanglements between two electron in adjacent dots for quantum based logic devices, sensors and communications.					
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FINAL REPORT
To
THE AIR FORCE OFFICE OF SCIENTIFIC RESEARCH

NONLINEAR OPTICS AND COHERENT OPTICAL CONTROL OF SINGLE ELECTRON SYSTEMS

AFOSR GRANT NO.FA9550-05-1-0150

GRANT PERIOD: 3/1/05 - 8/31/08

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Abstract

Work on this program was aimed at developing and understanding nano-optical structures with emphasis on developing quantum optical based devices. Specific work focused on semiconductor quantum dots. During this research period, a number of important discoveries were made as well as critical demonstrations of importance to future technology. The discoveries include the prediction and observation of spontaneous emission induced coherence and the unexpected control of nuclear field fluctuations through coherent electron spin trapping that reduced the nuclear fluctuations and increases the electron spin coherence time. Demonstrations include fast spin state initialization, coherent spin trapping, quantum dot tomography, and the Mollow absorption spectrum for neutral and negatively charged exciton. Future work will capitalize on this progress to demonstrate *deterministic entanglement* between the electron spin and a photon for quantum information transfer and entanglements between two electron in adjacent dots for quantum based logic devices, sensors and communications.

PUBLICATIONS

JOURNAL PUBLICATIONS

Most Significant Journal Publications

1. M.V. Gurudev Dutt, Jun Cheng, Bo Li, Xiaodong Xu, Xiaoqin Li, P.R. Berman, D.G. Steel, A.S. Bracker, D. Gammon, Sophia E Economou, Renbao Liu and L.J. Sham, "Stimulated and spontaneous optical generation of electron spin coherence in charged GaAs quantum dots," *Phys. Rev. Lett.* **94**, 227403 (2005).
2. Yanwen Wu, Xiaoqin Li, L. M. Duan, and D. G. Steel, D. Gammon, Lu Sham, "Density Matrix Tomography through Coherent Optical Rotation of an Exciton Qubit in a Single Quantum Dot," *Phys. Rev. Lett.* **96**, 087402 (2006).
3. C. Emary, Xiaodong Xu, D.G. Steel, S. Saikin, L.J. Sham, "Fast initialization of the spin state of an electron in a quantum dot in the Voigt configuration", *Phys. Rev. Lett.* **98**, 047401 (2007)
4. Yanwen Wu, Erik D. Kim, Xiaodong Xu, Jun Cheng, D. G. Steel A. S. Bracker, D. Gammon, Sophia Economou, and L. J. Sham, "Selective optical control of electron spin coherence in singly charged GaAs-Al_{0.3}Ga_{0.7}As quantum dots," *Phys. Rev. Lett.* **99**, p097402 (2007)
5. Xiaodong Xu, Yanwen Wu, Bo Sun, Qiong

- Huang, Jun Cheng, D.G. Steel, A. S. Bracker, D. Gammon, and C. Emary, L. J. Sham, "Fast Spin State Initialization in a Singly-Charged Quantum Dot," *Phys. Rev. Lett.* 99, 097401 (2007)
6. Xiaodong Xu, Bo Sun, Paul R. Berman, Duncan G. Steel, Allan Bracker, Dan Gammon, Lu J. Sham, "Coherent optical spectroscopy of a strongly driven quantum dot," *Science*, 317 pp 929-932 (2007).
 7. Xiaodong Xu, Bo Sun, Erik D. Kim, Katherine Smirl, P. R. Berman, D. G. Steel, A. S. Bracker, D. Gammon, and L. J. Sham, "A Single Charged Quantum Dot in a Strong Optical Field: Absorption, Gain, and the AC Stark, Effect" *Phys. Rev. Lett.* in press (2008).
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 9. Xiaodong Xu, Wang Yao, Bo Sun, Duncan G. Steel, Allan S. Bracker, Dan Gammon, L. J. Sham "Optically Controlled Locking of the Nuclear Field via Coherent Dark State Spectroscopy", submitted *Nature* (2008).

Additional Journal Papers

10. E.T. Batteh, Jun Cheng, Gang Chen, D.G. Steel, D. Gammon, D.S. Katzer, D. Park, "Coherent nonlinear optical spectroscopy of fluctuation quantum dots: evidence of coupling between quantum dots," *Phys. Rev. B* 71 p155327 (2005).
11. M.V. Gurudev Dutt, Yanwen Wu, Xiaoqin Li, D.G. Steel, D. Gammon, L.J. Sham, "Semiconductor quantum dots for quantum information processing : An optical approach", *Physics of Semiconductor*, José Menéndez and Chris G. Van de Walle, eds, Proceedings of the 27th ICPS, Flagstaff, AZ, American Institute of Physics Vol 772, p32-37, (2005).
12. Sophia E. Economou, Ren-Bao Liu, L.J. Sham, and D.G. Steel, "A Unified Theory of Consequences of Spontaneous Emission in a Λ System", *Phys. Rev. B* 71, 195327 (2005)
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14. M. V. Gurudev Dutt, Jun Cheng, Yanwen Wu, Xiaodong Xu, D. G. Steel, A. S. Bracker, D. Gammon, Sophia E. Economou, Ren-Bao Liu, and L. J. Sham, "Impulsive Stimulated Raman Control of Electron Spin Coherence in a Quantum Dot Ensemble", *Phys. Rev. B* 74, 125306 (2006).
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17. M.V. Gurudev Dutt, Jun Cheng, D.G. Steel, A.S. Bracker, D. Gammon and L.J. Sham, "A direct measurement of the trion relaxation and dipole decoherence rate: An application of high resolution coherent nonlinear optical spectroscopy," *Solid State Communications* 140, pp 374-377 (2006).
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20. Yanwen Wu, Erik D. Kim, Xiaodong Xu, Jun Cheng, D. G. Steel A. S. Bracker, D. Gammon, Sophia Economou, and L. J. Sham, "Selective Optical Control of Electron Spin Coherence in Singly Charged Quantum Dots via Oscillating Dark and Bright States," *Physical Review Letters*. 99, p097402 (2007).
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22. S. K. Saikin, E C. Emary, D. G. Steel, and L. J. Sham, "Adiabatic optical entanglement between electron spins in separate quantum dots," *Physical Review B* (2008).
23. S. K. Saikin, C. Emary, D. G. Steel, and L. J. Sham, "Adiabatic optical entanglement between electron spins in separate quantum dots". *Cond-mat/0802.1257*, submitted for publication.

INVITED CONFERENCE PAPERS

24. D.G. Steel, "Coherent optical manipulation of quantum dot spins: A path into quantum computing." *QELS* (2005).
25. Duncan Steel, "Application of Single Molecule Spectroscopy to the Study of Protein Folding Diseases," *International Symposium on Single Molecule Science*, 2006.
26. Duncan Steel, "Progress Towards Quantum

- Computing," *Frontiers in Quantum and Biological Information Processing*, Orlando, 2006.
27. Ren-Bao Liu, Sophia E. Economou, Lu J. Sham, and Duncan G. Steel "Theory on measuring electron spin decoherence time by nonlinear optical spectroscopy of single quantum dots "International Conference on the Physics of Semiconductors (2006).
 28. D.G. Steel, "Coherent Optical Control of Quantum Dots: Progress Towards Quantum Computing." NOEKS 2006
 29. Duncan G. Steel, "Quantum Dots for Quantum Computing," Rank Prize Fund, England 2007.
 30. D. G. Steel, *Fundamental Optical Processes in Semiconductors*, Montana, 2007
 31. D.G. Steel, "Atomic Physics in Artificial Atoms, Toward Coherent Manipulation of Single Electron Spins and Quantum Computing," International Conference on Quantum Information Rochester, NY 2007.
 32. D.G. Steel , "Optically Driven Electrons in Quantum Dots," *Frontiers in Optical Physics of Semiconductors*, Big Sky, Montana, 2007.
 33. D.G. Steel, "Optically driven quantum dot spins for quantum computing," NSF Workshop on Quantum Information Processing and Nanoscale Systems, Washington, September 2007.
 34. D.G. Steel, "Fast Optical Control of Spins in Quantum Dots for Quantum Technology Applications," Materials Research Society, Boston, Fall 2008.
 35. D.G. Steel, "Coherent Optical Control of Electron and Nuclear Spins in a Quantum Dot," American Physical Society, March Meeting, Pittsburg 2009
 40. Yanwen Wu, Erik D. Kim, Jun Cheng, Xiaodong Xu, D. G. Steel A. S. Bracker D. Gammon Sophia Economou and L. J. Sham, "Coherent Ultrafast Optically Controlled Rotation of Electron Spins in Charged Quantum Dots," QELS 2006 .
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 44. Xiaodong Xu, Bo Sun, Paul R. Berman, D. G. Steel, A. S. Bracker and D. Gammon L. J. Sham, "Coherent Optical Spectroscopy of a Strongly Driven Quantum Dot," QELS 2008.
 45. Erik D. Kim, Katherine Smirl, Yanwen Wu, A. Amo, Xiaodong Xu, and D. G. Steel, A. S. Bracker and D. Gammon, L. J. Sham, "On-Resonant Trion Rabi Oscillations and Spin Quantum Beats in a Singly Charged InAs Quantum Dot," QELS 2008.

CONFERENCE PRESENTATIONS

36. Xiaodong Xu, Jun Cheng, M. V. Gurudev Dutt, Yanwen Wu and D. G. Steel A. S. Bracker and D. Gammon, Renbao Liu, Sophia E. Economou and L. J. Sham, "Optically Stimulated and Spontaneously Generated Electron Spin Coherence in Quantum Dots" QELS/CLEO 2005
37. Jun Cheng, Xiaodong Xu, and D. G. Steel, A. S. Bracker and D. Gammon, L. J. Sham, "Spin-coherence-induced Hole Burning in Charged GaAs Quantum Dots," QELS/CLEO 2005.
38. Jun Cheng, Yanwen Wu, Xiaodong Xu, Dong Sun, D. G. Steel, A. S. Bracker D. Gammon, L. J. Sham, "Measurement of Electron Spin Relaxation in Charged GaAs Quantum Dots: Application of Nonlinear Optical Phase-Modulation Spectroscopy," QELS 2006 .
39. Sophia Economou, L. J. Sham, Yanwen Wu, and D. G. Steel "Theory proposal of electron spin rotation in a quantum dot," QELS 2006 .

EDUCATIONAL ACTIVITY

A number of students participated in the program as evidenced in the above publications. Three of the students have since graduated with a Ph.D and gone on to post-docs or permanent positions. Several new students have joined the group and are involved in the new program.

COLLABORATIONS

The work in the program is the result of an intense collaboration with Dr. D. Gammon at The Naval Research Laboratory supported by DARPA to develop quantum dot structures and spintronic based devices. In addition, the manybody theory component of the analysis of our findings is supported through our collaboration with Professor L.J. Sham (UCSD), supported by ARO, AFOSR and NSF.

SUMMARY OF FINDINGS

Nearly all of the research findings presented in this report have been reported in the annual reports. However, for completeness, we list the major developments, and we review a few of the most important results.

Introduction

The transfer of optical coherence to electronic coherence has featured prominently in current research as many new applications seeking to exploit the potential of this relatively new form of control. Electromagnetic induced transparency is one of the many areas where work is expanding as are related areas in quantum information and coherent optical control. While a few recent demonstrations have been reported in solid state systems such as impurity doped crystals and recently in silicon, the bulk of the work has been reported in atomic systems.

In this program, we built on our previous work demonstrating that quantum dot structures interact with coherent radiation similarly to atomic systems, unlike higher dimensional semiconductor systems that are characterized by complex many body interactions. We are using engineered structures that have had their electronic structure modified to create the ideal 3-level Λ system needed for demonstrations such as long coherence time for qubits in quantum computing, slow light and lasing without inversion.

The goal of the program is to demonstrate that the essential physical features can be seen in these systems and coherently controlled. Ultimately, the coherence time in these systems is limited by spin dephasing which can be very long. Given that these structures are created in III-V type material, developments in this area would naturally lead to easy integration with other optoelectronic devices.

Our approach to the study and manipulation of electron spin for application to spin based devices is based on the use of coherent nonlinear laser spectroscopy, coherent transient excitation and optical control, and the use of advanced semiconductor materials. Specifically, the electron spin, which would also correspond to the qubit for applications in quantum information science, is confined to a semiconductor quantum dot. Coherent control of the system is achieved by coherent optical excitation using the trion state as the intermediate state, thus allowing optical frequencies (eV) to be used to manipulate the spin states, separate by 10's of μeV . Materials are grown by MBE and further processed by lithography techniques by our collaborators, Dan Gammon and his group at NRL.

For quantum computing, a scalable architecture has been published by our collaborators (Lu J. Sham, UC-SD) based on individual qubits (electron spins) confined in adjacent quantum dots. Entanglement between spin in adjacent dots is accomplished by a modified optical RKKY (ORKKY) interaction resulting in a Heisenberg Hamiltonian coupling between the two spins.

Figure 1 shows the basic energy level diagram for InAs self-assembled quantum dots charged with a single electron and the corresponding optical selection rules for dots in an x-oriented magnetic field (Voigt profiles) resulting from addition of one electron. Two 3-level Λ -systems are produced. Relaxation between the two states is determined by spin relaxation and is known to be long, relative to the exciton relaxation time. The long relaxation time is expected to lead to long coherence times.

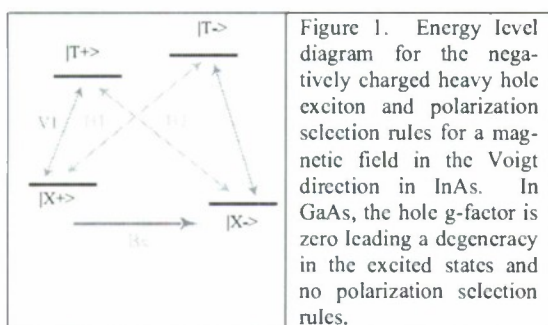


Figure 1. Energy level diagram for the negatively charged heavy hole exciton and polarization selection rules for a magnetic field in the Voigt direction in InAs. In GaAs, the hole g-factor is zero leading a degeneracy in the excited states and no polarization selection rules.

The usually forbidden optical transition between the trion state and the other spin state is allowed in the presence of a magnetic field in the x-direction (Voigt profile). Coherent optical control of the spin states is then enabled through a stimulated Raman two-photon (SR2P) pathway, shown by the red and green arrows.

Summary of the most important achievements:

Demonstration of selective optical control of electron spin

Discovery of spontaneous emission induced coherence.

First demonstration of qubit tomography in a solid.

Demonstration of fast initialization of the spin state of an electron in a quantum dot in the Voigt configuration.

Demonstration of impulsive stimulated Raman control of spin coherence.

Demonstration of the Mollow absorption spectrum in a single quantum dot showing GHz high speed switching.

Demonstration of AC Stark effect of a single electron transition that is important for future switching experiments.

Demonstration of coherent optical trapping and formation of the dark state in a single quantum dot.

Demonstration of optical Controlled Locking of the Nuclear Field via Coherent Dark State Spectroscopy

Discovery of hole-nuclear coupling and 5 orders of magnitude suppression of spin decoherence.

The technical portion of this report will feature just our most recent results.

Demonstration of the Trion Mollow Absorption Spectrum, Ultrafast Electron Switching and the AC Stark Effect in a Single Negatively Charged Dot

Because the excited state of a charged dot is a 3-body state, it is important to understand and demonstrate control of the physical properties of such a dot in the strong optical field regime, i.e. the light-matter interaction strength is much larger than the transition linewidth, under both resonant and nonresonant excitation. For an ideal two level atomic system, it has been shown theoretically and demonstrated experimentally that the strong coupling leads to new spectral features, such as Rabi side bands in the absorption, and strikingly, the amplification of a probe beam, which is known as the Mollow absorption spectrum (MAS)

The Mollow absorption spectrum was featured in the last report and was obtained on a neutral quantum dot. We have repeated those measurements now on a charged quantum dot and extended them to show the light induced level shifts of the AC-Stark effect.

The AC Stark effect plays a central role in QC architectures because it enables fast optical switching of energy states to bring states into or out of optical resonance. The results are especially important since a pub-

lication by another group in Europe failed to see the Mollow spectrum suggesting that negatively charged dots may not behave correctly allowing for Rabi oscillations. Fortunately, our observations show that in a correctly grown dot, the strong field behavior follows the standard theory.

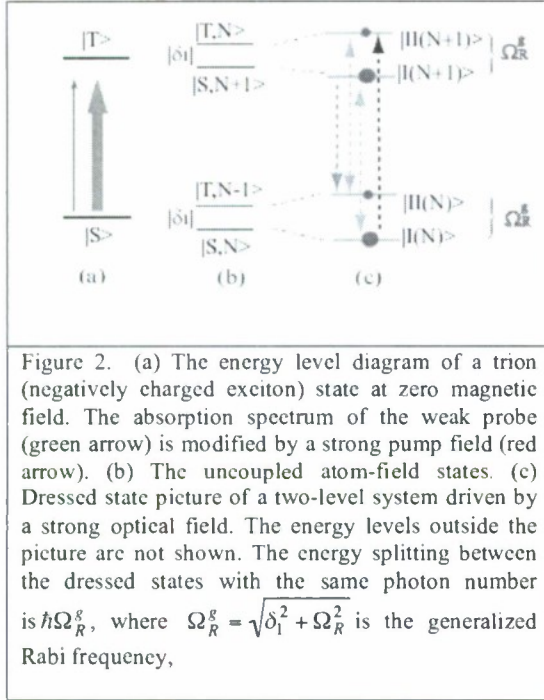


Figure 2 shows the basic approach of the experiment. A strong optical field drives the trion transition. A weak probe field then measures the resulting absorption spectrum. On the right of Fig. 2 is the dressed atom picture discussed in the previous report. When the pump detuning is large compared to the line width, level shifts are observed.

In Fig. 2, the relative size of the circles represents the relative population when the pump is red detuned. The transition centered at $\omega_1 + \Omega_R^g$ represents probe absorption (the purple dashed line in Fig. 2(c)), and the transition centered at $\omega_1 - \Omega_R^g$ is probe gain due to the population inversion of the dressed states (the red dashed line). The light blue lines indicate transitions where the probe

frequency is close to the pump frequency and the secular approximation fails. These can give rise to a dispersive lineshape.

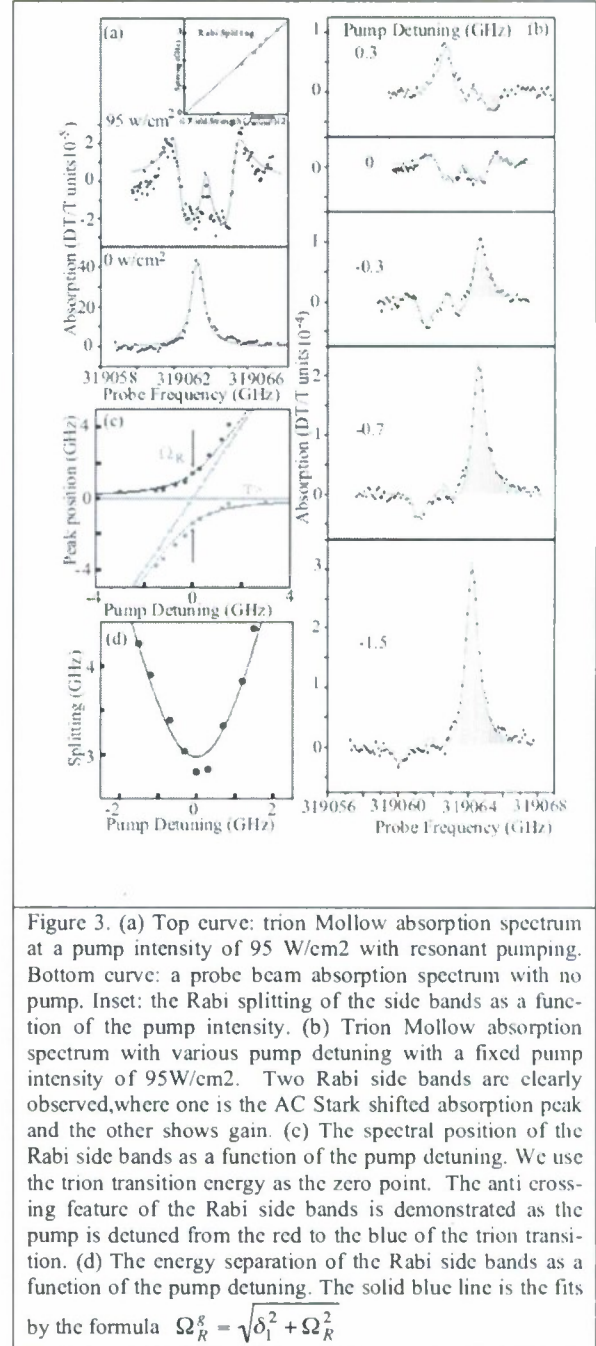


Figure 3 shows the essential results of this experiment. In Fig. 3a, the basic two side bands are revealed at high power with

their dispersive line shapes as predicted by theory (solid line) and separation given by the Rabi frequency. This key result shows the in spite of the manybody complexity of the trion state and the potential coupling to other states, the optical interaction follows the predictions of the 2-level model. The inset shows the linear dependence of the side band splitting on the optical field.

Figure 3b shows the absorption spectrum as a function of pump detuning with a fixed pump intensity of 95W/cm². Two Rabi side bands are clearly observed, where one is the AC Stark shifted absorption peak and the other shows gain.

In Fig. 3c we see the avoidance crossing due to the strong optical coupling to the system by examining the spectral position of the Rabi side bands as a function of the pump detuning. Figure 6d The energy separation of the Rabi side bands as a function of the pump detuning. The solid blue line is the fit by the prediction $\Omega_R^g = \sqrt{\delta_1^2 + \Omega_R^2}$.

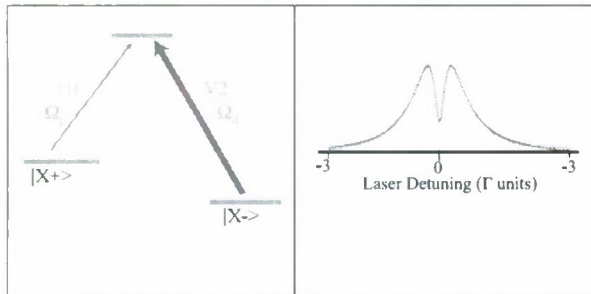


Figure 4. Left panel: 3 level structure for studying the dark state. Right panel: Theoretical line shape for absorption as a function of the detuning of the field nearly resonant with H1 as a function of detuning from H1. s

Demonstration of Coherent Spin Trapping, Formation of the Dark State, and Arbitrary Spin State Initialization

Using cw narrowband excitation, it is possible to excite just one of the trion states in Fig. 1. The resultant 3 level structure for

our experiment is shown in Fig. 4 (left panel). This is the same 3 level system used to optically initialize the system using optical pumping and spin cooling discussed in the previous report. When only V2 was on, the electron was optically pumped from the X- state to the X+ state.

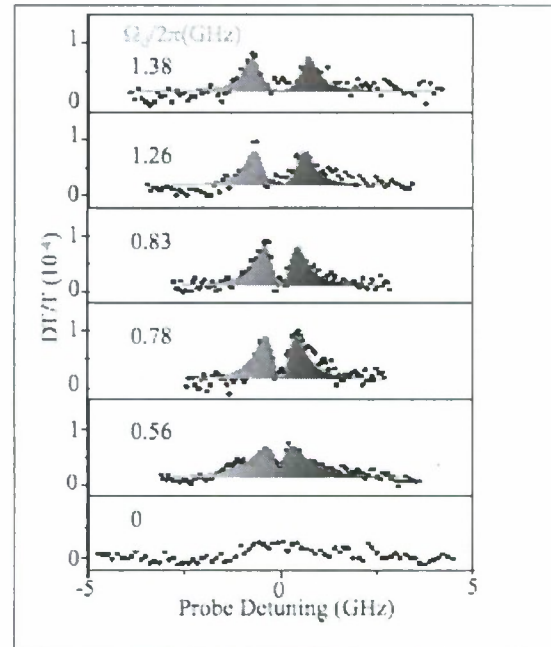


Figure 5. The dark state appears as the dip in the absorption spectrum as a function of probe detuning from the H1 resonance for various pump intensities, in units of the normalized Rabi frequency.

When a probe field simultaneously illuminates the system and is resonant with the other transition (H1), the system evolves, in steady state, to a dark state. This state is a superposition state of the two ground states given, in the field interaction picture, by

$$|Darkstate\rangle = \frac{\Omega_p |X_-\rangle + \Omega_d |X_+\rangle}{\sqrt{\Omega_p^2 + \Omega_d^2}}$$

and is manifest as a dip in the probe absorption spectrum when the probe is on the H1 resonance. The theoretical result is shown in the right panel of Fig. 4. The dip is evident even at low powers when the pump power is well below that needed to observe the onset of Autler splitting (cf last report.)

Figure 5 shows the experimental result for a number of different pump (V2) intensities (in units of the normalized Rabi frequency). The presence of the dark state dip is clear at intensities well below the onset of the Autler Townes splitting.

The dark state shows not only arbitrary spin state preparation but also further proof of the spin coherence since the dark state can not form in the absence of this coherence. The observation of the dark state is a necessary step in demonstrating the ability to line up the laser frequencies with the spin coherence bandwidth needed for spin rotation.

Demonstration of Ultrafast Coherent Control of the Electron Spin

In this program, we succeeded in achieving the basic demonstration of complete optical control of a single electron spin.

In these experiments, an external magnetic field of 5.5 T is applied perpendicular to the growth direction of the dot leading to the 4 level system shown in Fig. 1. Using the results of the previous report, a CW laser tuned to the negative trion resonance serves to initialize the electron spin into a pure state via optical pumping.

A single pulse is used to manipulate the initialized spin, and the result of the manipulation is read-out by the initialization beam as absorption.

For an on-resonant linear polarized pulse (Fig. 6, upper left panel), population is excited from the initialized spin to the negative trion states, which then spontaneously decays to the spin ground states with equal probabilities. Population that decays to the optically pumped spin state results in re-absorption of the CW probe beam, yielding a signal proportional to the amount of trion

population excited by the pulse. The re-absorption signal shows oscillations as the pulse amplitude is increased, reflecting trion Rabi oscillations.

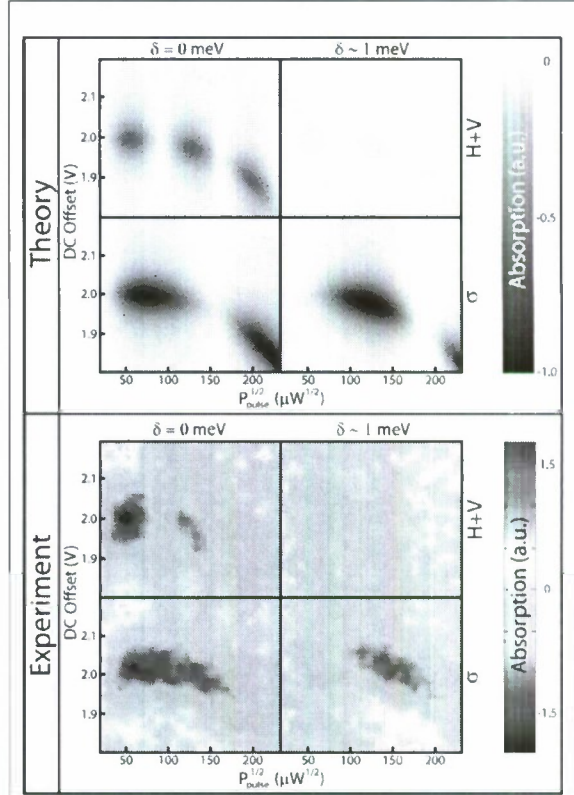


Fig. 6. Demonstration of coherent two-photon Rabi oscillations of the spin state. Theory (upper 4 panels) and experiment (lower 4 panels) Upper panels: Numerical simulations and experimental results of one-pulse electron spin control studies in a single dot as a function of detuning and incident polarization. For an on-resonant linear polarized pulse (upper left panel) population is excited from the initialized spin to the negative trion states, which then spontaneously decays to the spin ground states with equal probabilities. The signal shows oscillations as the pulse amplitude is increased, reflecting trion Rabi oscillations. Detuning the linearly polarized optical pulse reduces the amount of excited trion population, in turn reducing the re-absorption signal (upper right panel). In addition to generating trion population, an on-resonant circular polarized pulse (lower left panel) can drive transitions directly between the electron spin ground states via two-photon processes. As in the case of the linearly polarized pulse, detuning of the pulse suppresses the contributions from trion excitation. A two-photon spin Rabi oscillation is observed as the pulse amplitude is increased (lower right panel). There is good agreement between theory and experiment for low-to-moderate powers..

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dition to generating trion population, an on-resonant circular polarized pulse (lower left panel) can drive transitions directly between the electron spin ground states via two-photon processes, resulting in re-absorption of the CW beam. As in the case of the linearly polarized pulse, detuning of the pulse suppresses the contributions from trion excitation.

A two-photon spin Rabi oscillation is observed as the pulse amplitude is increased in both theory (upper panels) and experiment (lower panels). There is good agreement between theory (which includes the laser induced red shift as the power increases) and experiment for low-to-moderate powers. At higher power, the trion may be ionized or there may be increased coupling to tunneling states in the heterostructure. We believe this is due to a problem we are now addressing in dot growth, namely replacing Te doping with Si doping (Te is known to be more mobile during growth).

Coherent Optical Excitation and Readout of Electron Spin Coherence in Charged Quantum Dots

When the spin state is left in a coherent super position of $|X+\rangle$ and $|X-\rangle$, the state precesses in time, leading to oscillations in the absorption. Figure 7 shows the results of the measurement. This amounts to a rotation in the u-v plane of the Bloch sphere.

Numerical simulations and experimental results of two-pulse electron spin control studies in a single dot are again seen in the upper panels. By utilizing two detuned circular polarized pulses, the precession of the electron spin coherence generated by the first pulse is observed in the re-absorption of the CW beam as a function of the time delay between the pulses. The frequency of the precession, proportional to the Zeeman splitting of the electron spin states, may be controlled by varying the magnitude of the externally applied magnetic field.

The parameters for the numerical simulations were obtained from separately performed measurements of the electron and hole in-plane g-factors, showing good agreement with the experimental results in the lower panels.

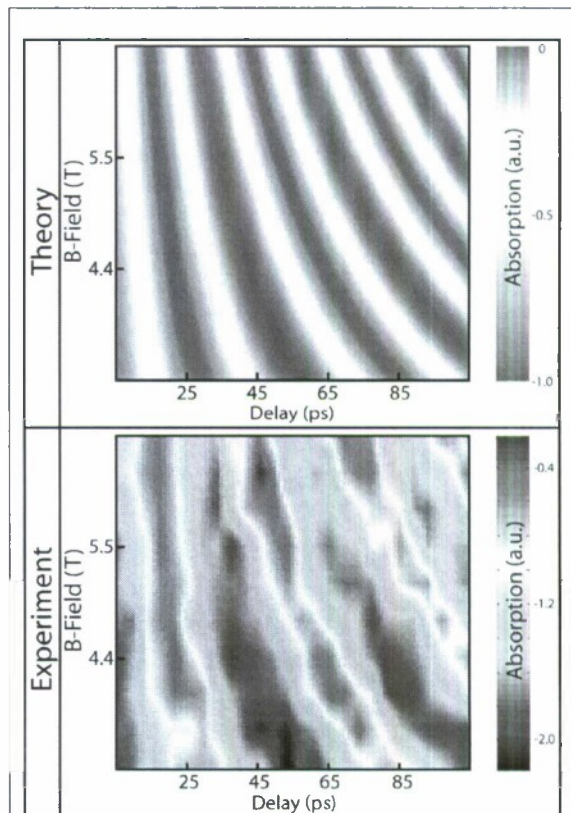


Figure 7: Numerical simulations (upper 4 panels) and experimental results (lower 4 panels) of two-pulse electron spin control studies in a single dot. By utilizing two detuned circular polarized pulses, the precession of the electron spin coherence generated by the first pulse is observed in the re-absorption of the CW beam as a function of the time delay between the pulses. The frequency of the precession, proportional to the Zeeman splitting of the electron spin states, may be controlled by varying the magnitude of the externally applied magnetic field. The parameters for the numerical simulations were obtained from separately performed measurements of the electron and hole in-plane g-factors.

Dynamic Control of Nuclear Spin Fluctuations

An unexpected result of our work has been the discovery that the optical field

could be used to control the nuclear polarization and reduce the nuclear fluctuations leading to an increase of the spin coherent time by over 2 orders of magnitude.

One of the acknowledged challenges to proposals using spin in III-V materials as the qubit is that the spin resonance and spin coherence are inevitably affected by the hyperfine interaction, and the dynamics of the spin can also actively affect the nuclear environment, for example through the Overhauser magnetic field.

Recent efforts have been made to enhance the electron spin coherence time (T_2^*) by suppressing the nuclear spin fluctuations in an ensemble of self-assembled QDs and in electronic gate confined double dot systems. However, simultaneous optical control of the nuclear spins and coherent control of the electron spin in a single dot has yet to be achieved and remains an outstanding bottleneck.

In this final period of the grant, we discovered we were able to optically suppress the fluctuations of the nuclear field in a singly charged quantum dot well below its thermal equilibrium, evident by a lower bound enhancement of the electron spin T_2^* . This optically controlled nuclear field locking via dark state spectroscopy fixes the nuclear field at a value determined only by the laser frequencies at fixed laser powers.

The nuclear field locking effect is found to result from an unexpected intrinsic hole-spin assisted dynamic nuclear spin polarization (DNP) feedback process. This is a simple and powerful method to enhance the electron spin coherence time without any complicated “spin echo” type techniques, where the coherence can only be transiently recovered at certain spin-echo times and the recovered coherence will only last for a

short duration equal to the non-enhanced T_2^* time.

We expect our results to provide the ability to reproducibly prepare the nuclear spin environment for repetitive control and measurement of single spins with minimal statistical broadening.

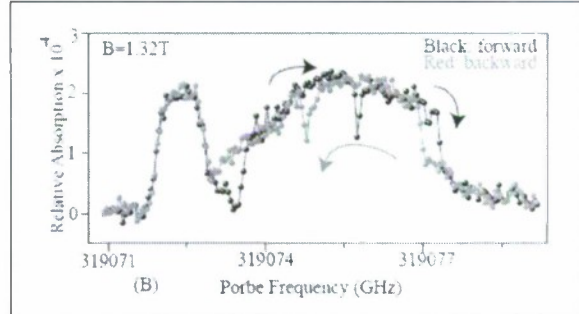


Figure 7. Hysteresis and irregular trion absorption line-shapes in the two beam experiments (the dip is the dark state from above) as a function of forward and backward probe scanning in the coherent spin trapping experiments described above.

The discovery started with the observation of hysteresis and irregular trion absorption line shapes in the two-beam experiments described above as a function of forward and backward tuning of the probe beam as shown in Fig. 7.

Yao and Sham developed a model that showed that the intrinsic hysteresis and distortion of the trion absorption profile was surprisingly not due to the electron because energy conservation requirements at this magnetic field made electron-nuclear spin flips a slow process. In fact, they showed the anisotropic term in the hyperfine interaction due to hole-coupling was the origin of the behavior and their theory beautifully reproduced the experimental features (Fig. 8).

Moreover, the model then predicted that that the coupling should lead to a reduction in the spin decoherence rate that increases with intensity. The dark state provided a convenient means to study the spin decoherence rate since the depth of the dark state

(reduction in absorption) increases with increasing spin coherence time. Figure 9a and 9b show this behavior, with Fig. 9a showing the decreasing decoherence rate with increasing power. The data shows a factor of 40 suppression of the nuclear spin fluctuations on the spin coherence.

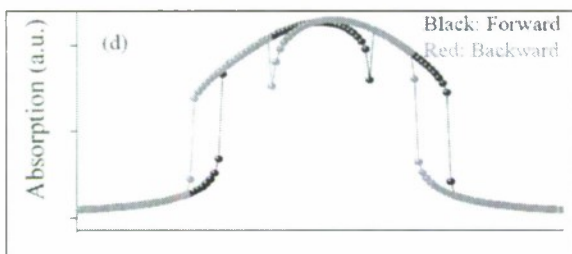


Figure 8. The model based on the anisotropic hyperfine interaction with the hole results in the observed hysteresis and distortion of the trion absorption. The sharp dip is the dark state.

The model shows that the nuclear spin precession is a function of the trion excitation level. As the nuclear field shifts to move the trion off resonance, the system works to restore the field to provide maximum trion excitation. This suggests that if two optical fields are used by locking the probe beam at the peak of the trion response in Fig. 9b (shown by the blue arrow), that we should then find a longer spin coherence time. Evidence to support this is seen in Fig. 9c where we have used a third optical field to scan the system. The dark state dip due to coherent spin trapping is much deeper, now, as anticipated. *A fitting of the data to the theory gives a spin decoherence rate that is instrument limited at 1 MHz, a factor of nearly 300 improvement.* The results are out for review at *Nature*.

Summary

This program has resulted in new understanding of importance to nonlinear optical nano-science, and has shown that nano-structures may be potentially viable structures for optically controlled quantum de-

vices. The studies have now evolved to studies of charged quantum dots that give rise to an energy level structure similar to that needed for many innovative applications such as for creating slow light devices and quantum logic gates.

The work has even shown control of the nuclear system which we are now examining to determine if it possible to transfer electron coherence to nuclear coherence for long term storage. This work has laid the foundation for our future studies of spin based quantum dots and more complex structures based on interacting quantum systems such as coupled quantum dots.

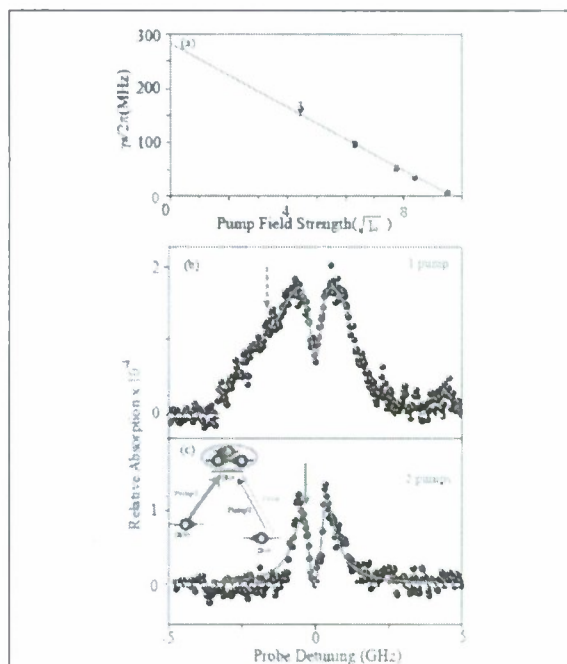


Figure 9. The model showed that the combined coherent coupling leads to a reduction in decoherence rate as the optical fields are increased in strength. Fig. 9a shows the dependence on power, Fig. 9b shows a typical data set used to extract the information for Fig. 9a. Figure 9c shows that locking the probe in Fig. 9b to the location of the blue line and scanning a 3rd optical field results in a greatly increased transmission at the dark state demonstrating that continuous locked fields can reduce the coherent rate by over 2 orders of magnitude (see text for discussion).